AECL

DEVELOPMENT, IRRADIATION TESTING AND PIE OF UMo FUEL AT AECL

by

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DEVELOPMENT, IRRADIATION TESTING AND PIE OF UMo FUEL AT AECL

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ABSTRACT

This paper reviews recent U-Mo dispersion fuel development, irradiation testing and post-irradiation examination (PIE) activities at AECL. Low-enriched uranium fuel alloys and powders have been fabricated at Chalk River Labs, with compositions ranging from U-7Mo to U-10Mo. The bulk alloys and powders were characterized using optical and scanning electron microscopy, chemical analysis, X-ray diffraction and neutron diffraction analysis. The analyses confirmed that the powders were of high quality, and in the desired gamma phase. Subsequently, kilogram quantities of DU-Mo and LEU-Mo powder have been manufactured for commercial customers. Mini-elements have been fabricated with LEU-7Mo and LEU-10Mo dispersed in aluminum, with a nominal loading of 4.5 gU/cm^3. These have been irradiated in the NRU reactor at linear powers up to 100 kW/m. The mini-elements achieved 60 atom% ^{235}U burnup in 2004 March, and the irradiation is continuing to a planned discharge burnup of 80 atom% ^{235}U. Interim PIE has been conducted on mini-elements that were removed after 20 atom% ^{235}U burnup. The PIE results are presented in this paper.

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1. Introduction

High density U-Mo dispersion fuel is being developed as a replacement for U_3Si and U_3Si_2 dispersion fuels in research and test reactors [1]. This international program has been undertaken to provide fuel with the higher densities needed to extend the use of low-enriched uranium (LEU) in research reactors, and to provide a fuel that is more easily reprocessed than silicide fuel [2]. Although both U_3Si and U_3Si_2 dispersion fuels perform well in AECL's research reactors, and in reactors that operate on LEU fuel supplied by AECL, the alternative UMo fuel offers an attractive option to close the back end of the fuel cycle should reprocessing of spent fuel be required. The UMo fuel development at AECL builds on the program that was previously used to successfully develop U_3Si and U_3Si_2 dispersion fuels for the NRU and MAPLE-type reactors [3]. A phased approach has been adopted, with the initial focus on UMo alloy development, powder fabrication and characterization. The second phase is focussed on prototype fuel fabrication, irradiation testing and performance analysis. Final scale-up and commercial implementation would follow after successful qualification and licensing.

This paper reviews recent U-Mo dispersion fuel development, irradiation testing and post-irradiation examination (PIE) activities at AECL.

2. Experiment

To obtain information on the performance and behaviour of U-Mo dispersion fuel under prototypical research-reactor conditions, we decided to irradiate mini-elements with a loading of 4.5 gU/cm^3. This is approximately 43% higher than the reference loading for the NRU or MAPLE reactors. The higher fissile loading in this fuel could be used to compensate for higher parasitic reactivity loads in experiments, to extend the fuel cycle (i.e., time between refuelling) or to reduce the refuelling requirements for a fixed operating cycle length, thereby reducing the fuel-cycle cost. Eight mini-elements were tested in NRU, four containing U-7wt% Mo, and four U-10wt% Mo particles dispersed in an Al matrix. Irradiating the two alloy compositions side-by-side allows for fuel performance comparisons under nominally identical operating conditions.

In addition, out-reactor tests were conducted to assess the thermal stability of the UMo dispersion fuel. Four mini-elements were heat-treated under vacuum (~10^{-6} torr) at 300°C, which is the maximum predicted fuel centreline temperature for fresh fuel under the irradiation conditions in NRU. The mini-elements were heat-treated for 1440 h, and diameter measurements were taken at 160 h intervals.

In a separate experiment, samples of the Al-U7Mo and Al-U10Mo fuel cores, about 25 mm long, were heat-treated in a tube furnace at 400 and 500°C for 15 h under vacuum (~10^{-6} torr). After the heat treatment, the samples were visually examined, photographed and the dimensions and weights of the samples were measured. Subsequently, samples were heated to 700°C in vacuum and argon, to evaluate the fuel behaviour under extreme high-temperature conditions. Metallographic examinations and neutron diffraction analyses (NDA) were performed to assess the microstructure and phase changes.

3. Fuel Fabrication

The Al-U7Mo and Al-U10Mo mini-elements were fabricated at the Chalk River Labs (CRL). Details of the fuel core compositions are given in Table 1.

Table 1. Nominal U-Mo Mini-Element Fuel Core Composition.

Al–U-Mo Mini-element Fuel Core Loading	**Al - 71.2 wt % U7Mo**	**Al - 72.4 wt % U10Mo**
Core density, g/cm^3	6.79	6.91
Core mass, g	24.0	24.5
U-Mo mass, g	17.1	17.7
U mass, g	15.9	15.9
U density in fuel, g/cm^3	4.5	4.5
Linear loading, gU/cm	1.4	1.4
Mass ^{235}U/cm, g/cm	0.3	0.3

A description of the typical mini-element manufacturing process and the dimensions are given elsewhere [3]. The U-Mo alloys were made by vacuum induction melting the constituent metals, followed by casting. Cast billets were heat-treated under vacuum at 900 °C for 72 h then quenched to preserve the metastable γ phase. Powders were produced from the bulk alloys using a proprietary process at Chalk River. The powder was characterized using optical and scanning electron microscopy (SEM), chemical

analysis, and X-ray and neutron diffraction analyses (NDA). The NDA pattern (Fig. 1) shows the as-fabricated powder retained the γ (U, Mo) phase. UMo powder was mixed and blended with Al powder, extruded into cores, and fabricated into mini-elements.

In 2003, several kg quantities of DU-7Mo and LEU-7Mo powder were produced on a commercial basis at AECL. Fig. 2 shows the typical morphology of the LEU-Mo powder produced at Chalk River.

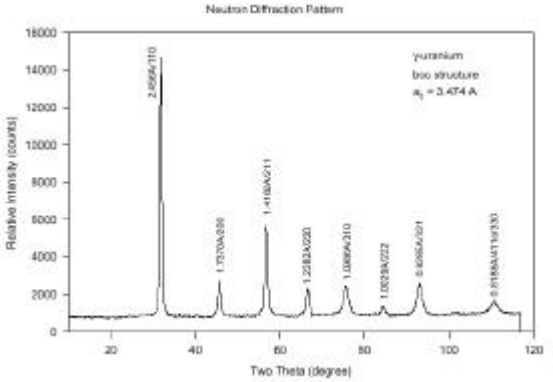


Fig. 1 Neutron diffraction pattern of as-fabricated UMo powder.

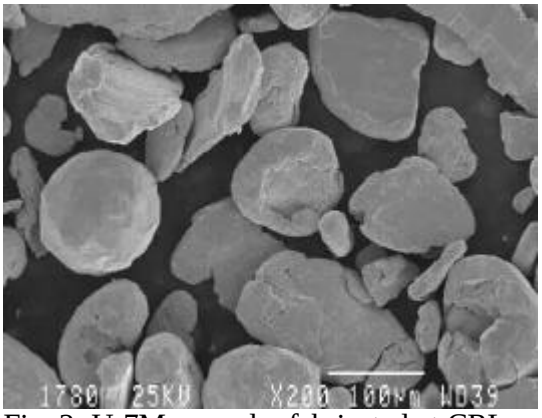


Fig. 2 U-7Mo powder fabricated at CRL.

4. Irradiation History

The UMo fuel test irradiation started in the NRU reactor on 2003 May 12, and Phase I (to 20 at% burnup) was completed on 2003 July 14. After each phase, a string of mini-elements was removed for interim examination, replaced with dummies, and the test rod was returned to the reactor to continue the irradiation. Table 2 shows the irradiation history to the end of 2004 September. The nominal D_2O coolant inlet temperature was ~ 35°C; conductivity was < 0.3 µS/cm; and pH was 5.5 - 6.5. The coolant velocity was typically ~8 m/s over the heated element surfaces. The power history for the UMo fuel irradiation is shown in Fig. 3. During the first two phases of the irradiation, the element linear rating was limited to maximum 100 kW/m. In subsequent phases the power was allowed to decline with burnup, as would be typical in service.

Table 2: Irradiation of UMo Fuel in NRU

In/Out of Reactor	atom% burnup	
YY/MM/DD	U-10%Mo	U-7%Mo
In: 03/05/12	0	0
Out: 03/07/14	20.04	20.20
In: 03/09/16	20.04	20.20
Out: 03/11/12	39.32	40.40
In: 04/01/22	39.32	40.40
Out: 04/03/29	59.72	60.62
In: 04/07/30	59.72	60.62
In: 04/09/15	68.00	69.19

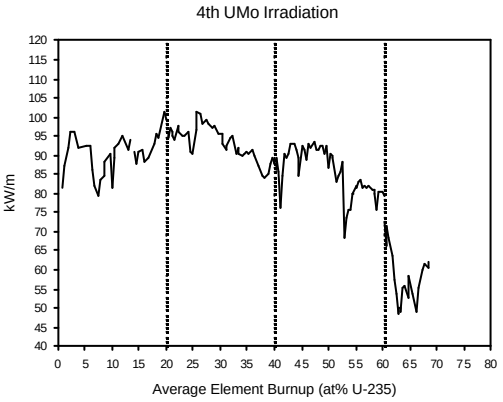


Fig. 3 Power History of UMo Fuel.

5. Post-Irradiation Examination

Underwater visual examinations were conducted after each phase of the irradiation and the fuel was found to be in good condition with no evidence of defects, deformation, or oxide spallation.

Gamma-scans were collected after 20 at% burnup. The mini-elements were rotated about the longitudinal axis and scanned with a lithium-drifted germanium detector.

Immersion density was measured in the as-discharged condition, and again after the surface oxide layer was chemically stripped using a hot solution of 79% distilled water, 20% phosphoric acid and 1% nitric acid. The results were used to calculate the core swelling.

The dimensions of the mini-elements were measured at five axial locations. The cladding diameter was measured between opposing pairs of fins.

Fuel samples were extracted from the midplane, dissolved in nitric acid, and analyzed using thermal ionization mass spectrometry (TIMS) for U isotopic abundance. For the uranium analysis, the U was separated from the fuel matrix by HPLC. The burnup was calculated from the pre- and post-irradiation isotopic ratios.

Samples were cut from the midplane (transverse) and ends (longitudinal) of the mini-elements, then mounted and polished for optical metallography (OM). Radial slices were cut from the transverse samples, then mounted adjacent to as-fabricated core samples for scanning electron microscopy. The concentration profiles for the major phases in the fuel were determine using Wavelength Dispersive X-Ray (WDX) analysis on the SEM. Line scans and large area X-ray maps were collected to assess the partitioning of the constituent elements.

6. Results

6.1 PIE Results

Visual examination revealed the usual grey surface oxide layer over the heated surfaces, with a darker oxide on the endplug region (Fig. 4). No unusual features were observed.

The gamma scans (Fig. 5) show a relatively uniform distribution of fission products along the fuel cores. This is typical of LEU mini-elements that are irradiated near the axial midplane of NRU where the flux profile is relatively flat. The step change in activity at each end corresponds to the drilled out annular region where the end plug is inserted into the fuel core. The higher activity at the end of the solid core and drilled out section is consistent with end-flux-peaking effects. Uranium isotopic analysis results are shown in Tables 3. The calculated burnup, 20 at% U-235, is in good agreement with the physics code predictions.

Table 3. Uranium atom ratios in UMo Fuel after Phase 1 Irradiation

Sample I.D.	235/238	234/238	236/238
U-10%Mo 49-1B	0.1974 (1)	0.001502 (4)	0.01289 (4)
U-7%Mo 59-1B	0.1990 (1)	0.001511 (5)	0.01294 (3)

*the error for one standard deviation is given in parenthesis, corresponding to the last digit

The diameter measurements indicated that the cladding strain was ~ 3% at the midplane of both mini-elements, and ~ 2% at the ¼- and ¾-plane locations. Immersion density measurements indicate that the fuel cores swelled by 5.7 vol% for the U-10Mo and 5.9 vol% for the U-7 Mo fuel. The swelling in the UMo fuel is consistent with the microstructural changes discussed below. Metallographic samples showed ~ 2.6-2.9% diameter increase (midplane), consistent with the average immersion density swelling.

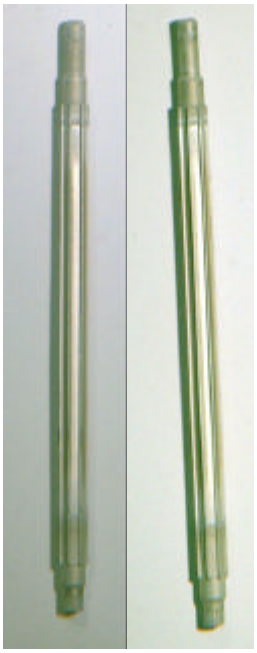


Fig. 4 UMo mini-elements after 40 at% BU

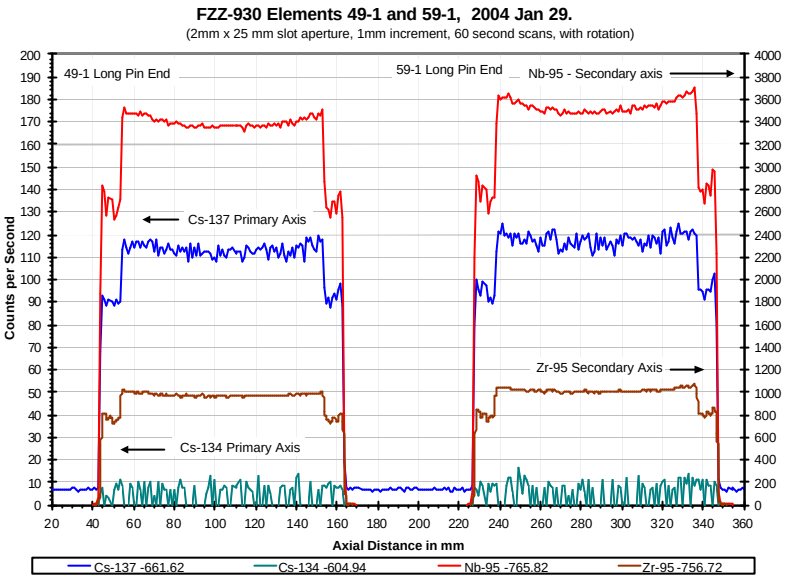
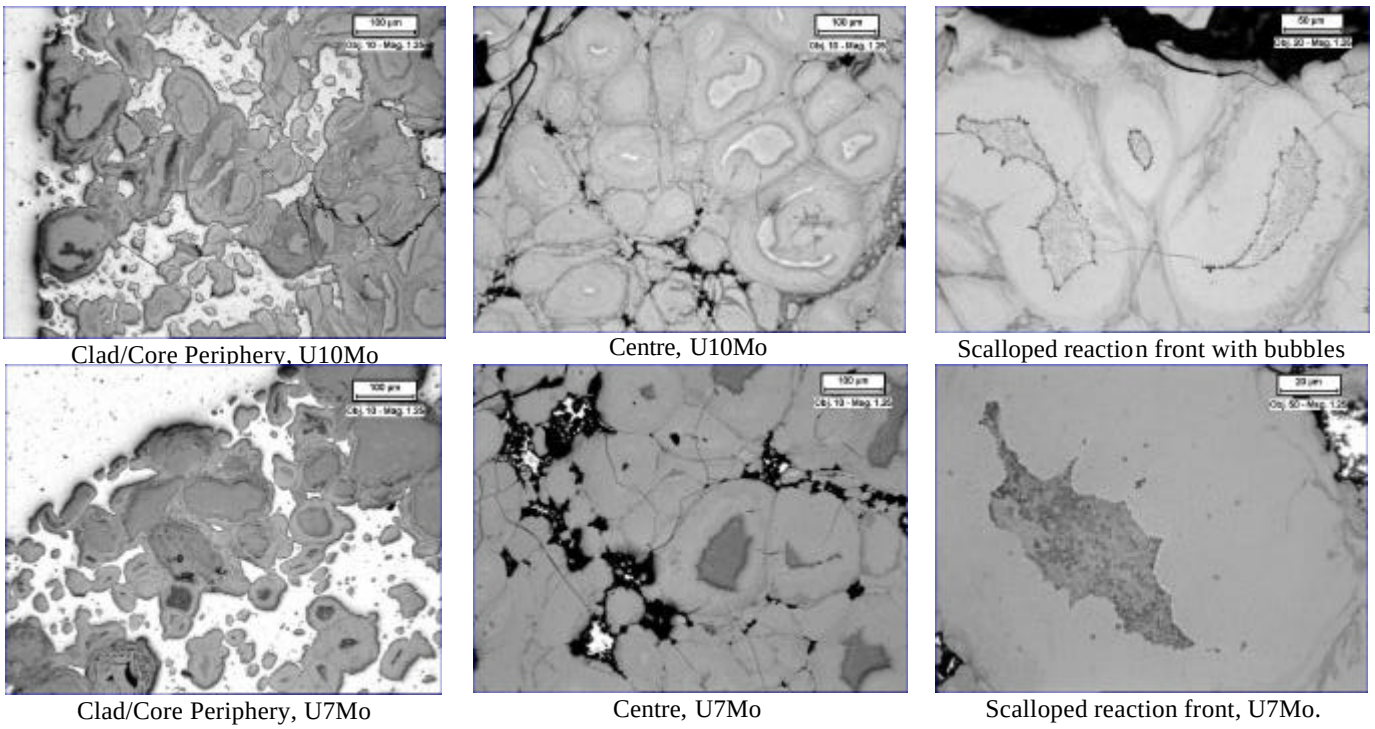


Fig. 5 Gamma scans of U-7Mo and U-10Mo mini-elements after 20 at% burnup.

The OM examination revealed signification reaction between the fuel particles and aluminum matrix. Fig. 6 shows the microstructure of the UMo fuel. At the periphery, evidence of the Al matrix (white) and original discrete UMo particles (dark grey) remained, but thick reaction layers (light grey) had grown at the fuel-particle/matrix interface. At the centre region, interaction between the UMo and Al almost fully consumed the Al matrix, and the U-Mo-Al reaction product has become the major phase, surrounding residual kernels of the UMo particles. As seen in Fig. 6, the reaction front at the UMo kernel interface has a scalloped profile, and in the U10Mo fuel this interface line was decorated with small bubbles, possibly containing fission gas. A similar scalloped reaction front was observed in the U7Mo fuel but fewer bubbles were evident. The SE images in Fig. 7 more clearly show the extent of reaction between the UMo and Al, from periphery to centre. The swollen fuel-particle boundaries are well delineated even where the fuel has completely reacted and consumed the Al. Voids were observed in the centre region, in part due to pull-out during sample preparation, despite vacuum impregnation. Radial cracks were also observed out to about mid-radius.

Figures 8 and 9 show the SE images and large area X-ray maps for U, Mo and Al in the two fuels. The Al maps show that the matrix was fully consumed beyond the mid-radius

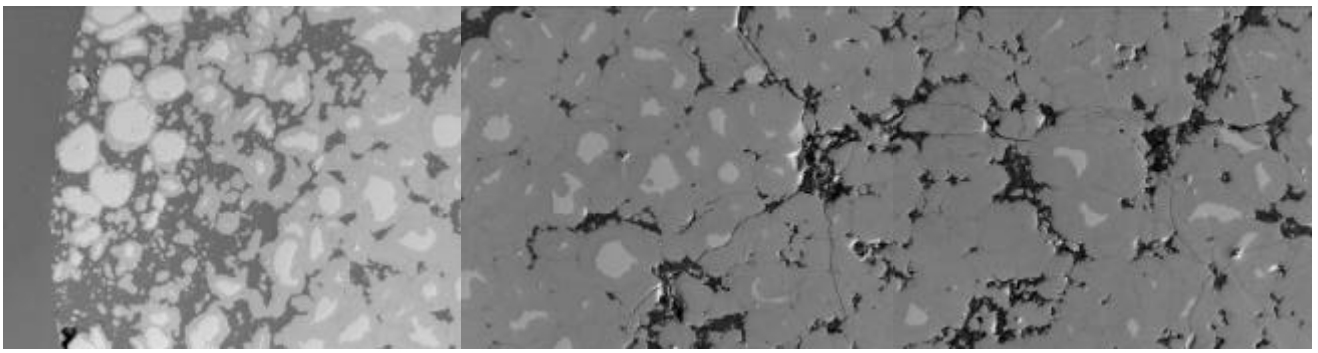
region of the U10Mo sample (Fig. 8), while residual islands of Al could be seen in the U7Mo sample (Fig. 9). Interdiffusion has homogenized the composition in the central region in both samples, with higher concentrations of U and Mo detected only in a few residual kernels.



Clad/Core Periphery, U10Mo | Centre, U10Mo | Scalloped reaction front with bubbles

Clad/Core Periphery, U7Mo | Centre, U7Mo | Scalloped reaction front, U7Mo.

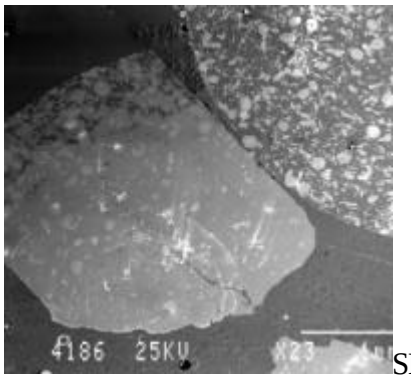
Fig. 6 Optical micrographs of U10Mo and U7Mo fuels after 20 at% burnup.



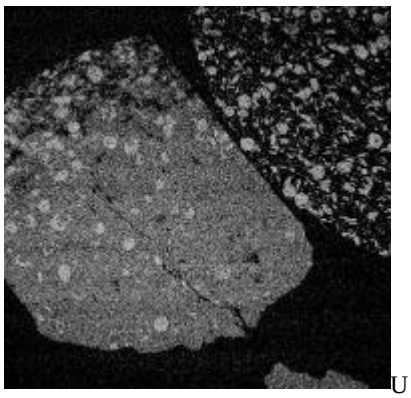
5013 Periphery, U7Mo 5014 Midradius, U7Mo 5015 Near Centre, U7Mo

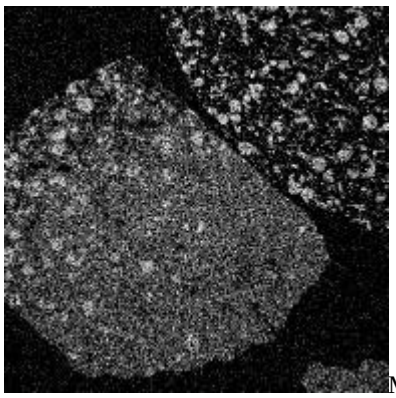
Fig. 7 SEM SE images of U7Mo fuel after 20 at% burnup.

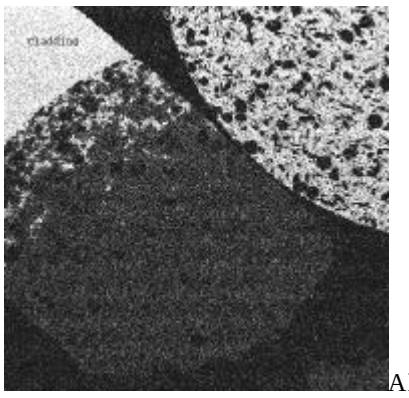
Semi-quantitative X-ray line scans (Fig. 10) show the U, Mo and Al distribution with the fuel. The U distribution appears relatively homogeneous within the kernel, and within thick interfacial layers. The average composition of the U-Mo-Al reaction product was found to be between 77 and 82 at% Al, with a U/Mo ratio ~ 3, corresponding to phases denoted as (U, Mo)Al_3 and (U, Mo)$Al_{4.4}$ [4]. Similar compositions were reported in UMo-Al diffusion couples [5] and in fuel samples irradiated at high temperature [6]. Both UAl_3 and UAl_4 have lower densities than the starting material [7].



SEI

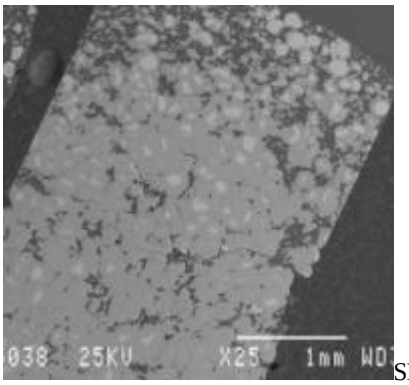

U


Mo

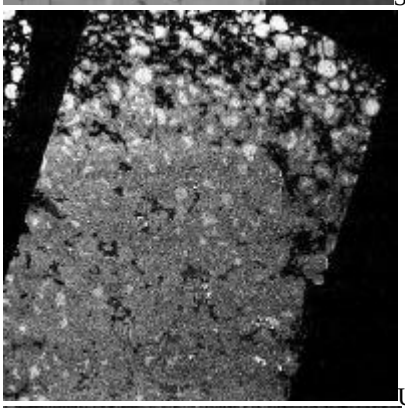


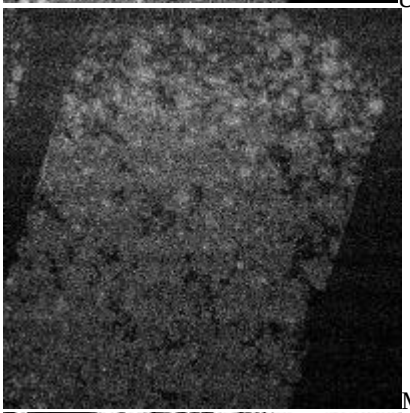
Al

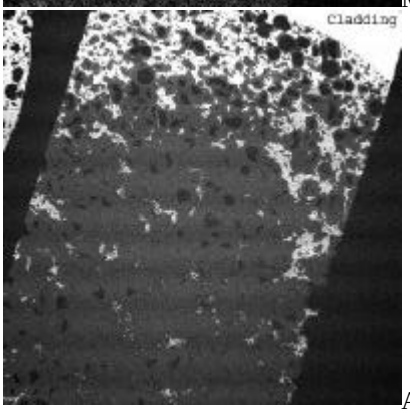
Fig. 8 SE image and large area X-ray maps of U, Mo and Al distribution in the Al-U10Mo fuel core. Each image shows the 20 at% BU sample (lower left), and an adjacent as-fabricated sample (right).



SEI


U


Mo



Al

Fig. 9 SE Image and large area X-ray maps of U, Mo and Al distribution in the Al-U7Mo fuel core. Each image shows the 20 at% BU sample (right), and an adjacent as-fabricated sample (top left).

6.2. Thermal Compatability Assessments

The 4 mini-elements that were vacuum annealed to 300°C for 1440 h appeared to be unchanged from the as-fabricated condition. The dimensional and immersion density measurement taken at 160 h intervals showed negligible changes, indicating that the UMo fuel is stable under these test conditions. Fuel core samples that were heated to 400°C for 15 h also showed negligible volume changes, but after 15 h at 500°C, the U-7Mo core diameter increased by 5.8%, and the U-10Mo core diameter increased by 7.9%. The core samples heated to 700°C for 1 h showed diameter increases ($\Delta d/d$) of ~ 20-22%. The cores were analysed by NDA, and the results are shown in Table 4. NDA patterns and micrographs of the fuel sample annealed to 700°C are shown in Fig.11. The Al was completely consumed and only γ-U (no α) and UAl_3 were identified. Several unidentified peaks were observed in the diffraction pattern from the 700°C samples; these could not be matched with known phases in the materials database so the weight percentage results shown in Table 4 are normalized.

Table 4. Thermal Compatability of Al-UMo Fuel and NDA results (wt% of each phase detected).

ID	**Composition**	**T, °C**	**Time, h**	**Dd/d, %**	**Al**	**g-U**	**a-U**	**UO2**	**UAl3**
B	Al-U10Mo	400	15	0.16	26	55	13	6	n.d.
A	Al-U10Mo	500	15	7.87	21	38	25	5	11
D	Al-U7Mo	400	15	0	25	44	26	4	n.d.
C	Al-U7Mo	500	15	5.83	22	34	26	3	15
"Vac"	Al-U10Mo	700	1	20.2	n.d.	14	n.d.	n.d.	86
"Ar"	Al-U10Mo	700	1	21.9	n.d.	14	n.d.	n.d.	86

7. Discussion

The UMo fuel core swelling is significantly higher than observed in comparable silicide fuels where, under equivalent irradiation conditions, 5-6 vol% swelling is typically observed after ~ 80 at% burnup [7]. At the low burnup achieved in NRU, the contribution of fission products to swelling is expected to be negligible. The swelling is attributed to the interaction of U-Mo-Al and the formation of lower density reaction products. The interdiffusion of UMo and Al is clearly temperature dependent, forming thick reaction products at the hot centre of the fuel core and thinner interfacial layers at the cooler periphery. The microstructural evolution at mid-radius appears to be consistent with that observed in a high-power test irradiation (FUTURE) in the BR2 reactor [6] and in out-reactor annealing tests [4,5]. The UMo fuel was irradiated to 33% burnup in BR2 and swelled by 13 vol%. Considering the higher loading (8.4 vs. 4.5 gU/cm^3) and the higher burnup achieved (33 vs. 20 at%), the core swelling in NRU seems consistent with the trend. The FUTURE experiment was prematurely terminated because of excessive swelling of the plate fuel, and PIE revealed the linking up of voids, which could lead to break-away swelling and pillowing of plate-type fuel. Voids were observed at the Al matrix/reaction layer interface in both mini-elements, which can be attributed to the Kirkendal effect associated with the diffusion of Al and movement of vacancies [6]. However, similar linking up of voids and break-away features were not observed in the mini-elements. The mechanical restraint offered by the finned cladding in our cylindrical pin designs helps to restrain swelling of the fuel core.

The combined effect of voids and the large volume fraction of reaction product in the

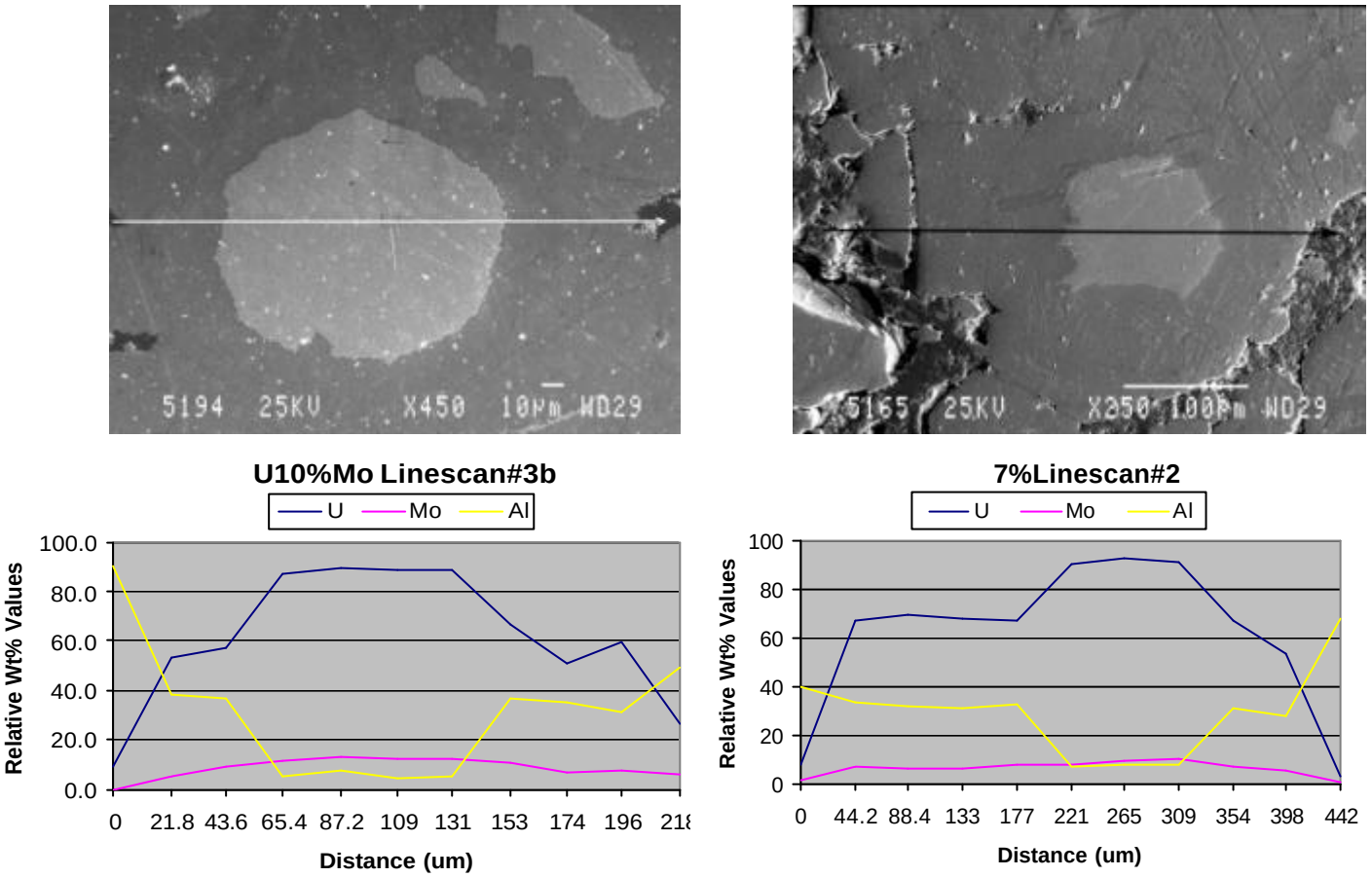


Fig. 10 Semi-quantitative line scans of Al-U10Mo (left) and Al-U7Mo (right).

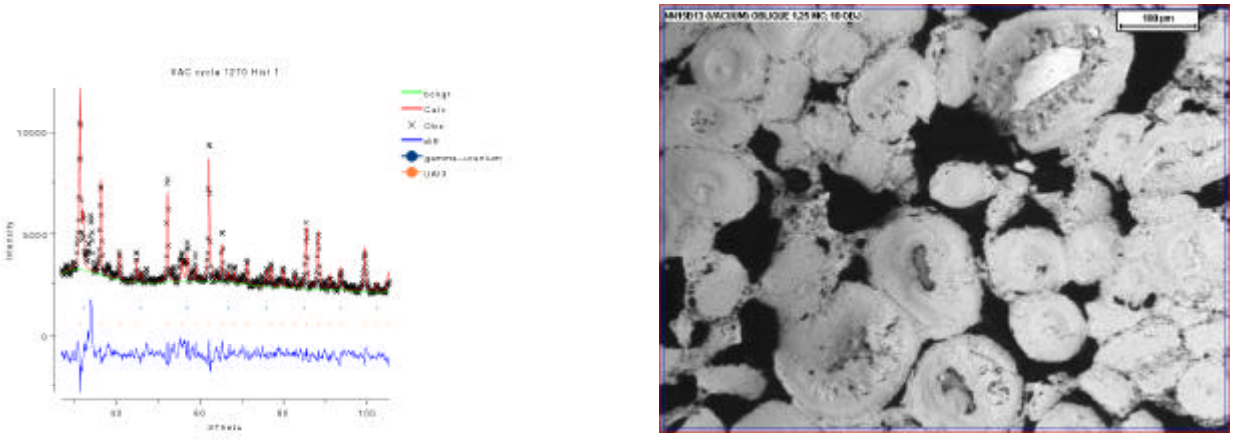


Fig. 11 Neutron diffraction analysis pattern and micrograph of Al-U10Mo fuel annealed at 700 °C for 1 h.

central region will reduce the thermal conductivity of the core as the lower conductivity reaction product replaces the high conductivity Al matrix. At constant power, the net effect would be an increase in fuel central temperature. However, the fuel temperature during irradiation is a complex function of power with time and the thermal properties of the core. The negative feedback effect of phase change on fuel thermal conductivity further complicates the calculation. Hayes has treated the issue in detail and is developing a model to accommodate these changes in plate-type fuel [8]. A similar code is not yet available for pin-type fuel. Further work is required to assess the maximum fuel temperature achieved during irradiation in NRU.

8. Conclusions

Al-U10Mo and Al-U7Mo dispersion fuel elements have been fabricated at Chalk River and successfully irradiated in the NRU reactor to burnups above 70 at% U-235 depletion. Irradiation to 20 at% burnup, at linear element ratings up to 100 kW/m, results in fuel core swelling of approximately 6 vol%. At low burnup, the contribution of fission products to swelling is expected to be negligible. PIE shows that the core swelling is largely the result of U-Mo-Al interaction, which forms a reaction product with lower density than the as-fabricated dispersion. Porosity has been observed in the fuel core, but this does not appear to cause unacceptable swelling or blistering of the mini-elements under the conditions tested. The mini-elements are expected to achieve the design burnup of 80 at% U-235 by the end of 2004.

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